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Effect of Low Ambient Temperature on Emissions and Electric Range of Plug-In Hybrid Electric Vehicles

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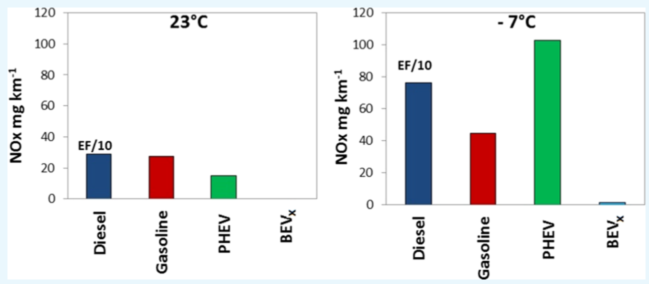
Supporting Information

ABSTRACT: Plug-in hybrid electrical vehicles (PHEVs) are generally considered to be a cleaner alternative to conventional passenger cars. However, there is still very limited information available regarding criteria pollutant emissions from these vehicles. This paper shows, for the first time, the emissions of criteria pollutants, unregulated pollutants, and CO_2 and also electric range from two very different PHEVs, one Euro 6 parallel plug-in hybrid and one range-extended battery electric vehicle (BEVx), applying the new world harmonized light-duty test procedure at ambient temperatures equal to 23 and −7 °C. The impact of using a cabin air heating system on vehicle electric range and emissions at cold temperature has also been studied. Cold ambient temperatures and, to a larger extent, the use of heating systems have been shown to lead to a pronounced negative impact on emissions and shorter electric ranges. Results also show that modern PHEVs can emit similar, or even higher, levels of pollutants (e.g., particle number) as Euro 6 conventional gasoline and diesel vehicles.



1. INTRODUCTION

Vehicle exhaust emissions are of general concern as they are among the main contributors to urban air pollution and to climate change. Vehicles emit air pollutants such as NOx, volatile organic compounds (VOCs), NH_3, and fine particles[1−3] and also greenhouse gases (GHG)—mainly CO_2, N_2O, and CH_4.[4] Plug-in hybrid electrical vehicles (PHEVs) (also known in the European Union (EU) as off-vehicle charge hybrid electric vehicles—OVC-HEVs) are presented as a sustainable mobility alternative to reduce the vehicle emissions. PHEVs are vehicles equipped with an internal combustion engine (ICE), an electric motor, and a rechargeable electric energy storage system (REESS) that can be directly charged from the electric grid. PHEVs represent a technical compromise between pure-electric vehicles (PEV) and the conventional vehicles. They offer drivers the same range as conventional vehicles while providing the environmental benefits of pure-electric vehicles (PEVs), such as the absence of exhaust pollutant emissions during electric operation and reduction of GHG emissions. Owing to these features, PHEVs will be allowed to circulate in low-emission zones that have been defined aiming at improving urban air quality.

Faria et al. have recently showed that GHG emissions would be substantially reduced by PHEVs used under the present EU energy generation system.[5] According to Plötz et al. (2017), these vehicles could be charged using renewable electricity, and they are seen as a proxy to meet the transport GHG reduction targets.[6] Moreover, the possibility to lean on electrical energy for transportation purposes limits the energy dependency on fossil fuels, which is of extreme importance for the EU.[7]

The European Environmental Agency (EEA) presents every year a comprehensive emission inventory that includes vehicle emissions. This inventory is often used for life-cycle assessments and model studies in the EU, and yet, in the latest EEA emission inventory, there are no emissions listed for PHEVs. The document states that they will have very low, but nonzero, emission rates, and since the number of these vehicles is currently very low, their emissions could be neglected for the time being.

Previous works reported that hybrid electric vehicles present enhanced energy efficiency and lower pollutant emissions compared to conventional vehicles,[8,9] but we have recently reported that criteria pollutant emissions from a Euro 5 PHEV can be similar to those measured from conventional vehicles.[10] Yuksel et al.[11] have recently shown that life-cycle plug-in electric vehicle CO_2 emissions depend on the assumed electricity grid mix, driving patterns, and ambient temperature. Low ambient temperature has been proven to reduce PHEVs' and PEVs' electrical range,[11−14] to increase exhaust emissions from hybrid electric vehicles,[8] and to reduce environmental benefits from PEVs compared to conventional gasoline

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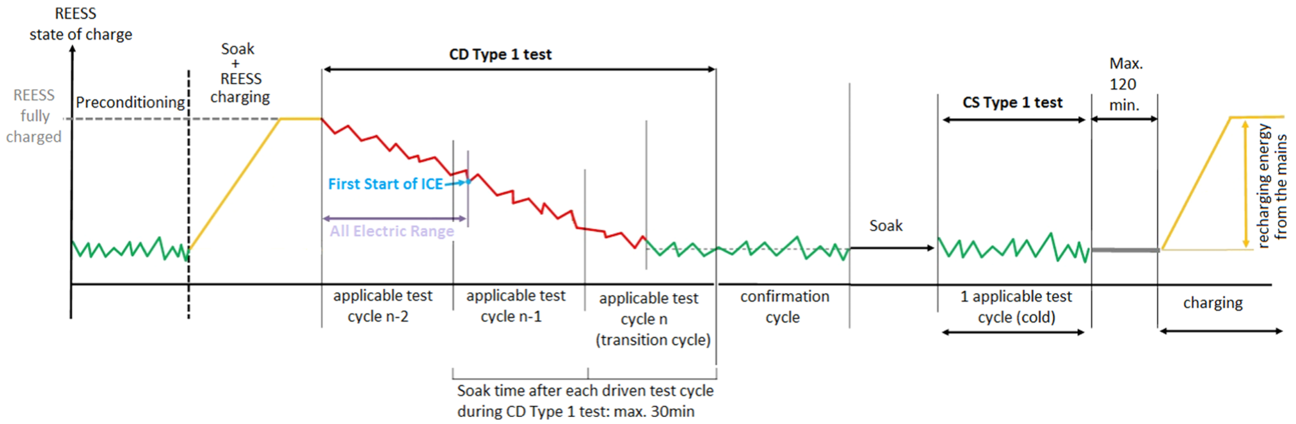

Figure 1. Test sequence for OVC-HEVs according to option 3, including charge-depleting (CD) and charge-sustaining (CS) type 1 tests, as described in Annex 8-Appendix 1 of GTR-15.

vehicles.[15] Same as for conventional vehicles, PHEVs' exhaust emissions may be a concern during the winter season, which is already associated with high pollution episodes.[16,17]

United Nations Economic Commission for Europe (UNECE) Regulation 101[18] was used to test the hybrid electrical vehicles until very recently. This procedure was heavily criticized for its favorable testing assumptions for PHEVs since it was not representative of real-world operations. That procedure has been replaced by the world harmonized light-duty test procedure (WLTP).[19] The WLTP, developed to be more representative of real-world driving conditions than the former type-approval, has been used for type-approval of light-duty vehicles (LDVs) in the European Union since September 2017.[19] Since PHEVs are gradually taking over a large fraction of the global vehicle market,[20] and they are considered to be a cleaner alternative to conventional gasoline and diesel passenger cars, it is of major importance to evaluate their emissions to be able to anticipate their impact and share in the total emissions from the transport sector.

In this context, we have studied two modern PHEVs applying the WLTP. The vehicles presented two different architectures, one parallel—which uses both ICE and electric motor to propel the vehicle—and one series—which uses an auxiliary power unit (APU) as generator, supplying electricity to the electric engine, which provides the energy needed to propel the vehicle. We also have evaluated how other real-world conditions, such as the cold ambient temperature and the use of air heating system, can impact the emissions and electric range. In this work, we aim at highlighting the importance of the emissions of regulated (total hydrocarbons (THC), NOx, CO, particle number (PN)) and nonregulated (NH_3 and N_2O) pollutants from PHEVs in comparison to conventional vehicles, as well as the strong negative effect that cold ambient temperature can have on PHEV's emissions and vehicle electric range.

2. EXPERIMENTAL SECTION

2.1. Test Vehicles. Two gasoline PHEVs, one Euro 6 with parallel configuration (hereinafter PHEV) and one US range-extended battery electric vehicle (BEVx) equipped with a range extending APU (hereinafter BEVx), were tested on a chassis dynamometer in the Vehicle Emission Laboratories (VELAs) at the European Commission Joint Research Centre (EC-JRC) Ispra, Italy.

The PHEV was a European Euro 6a vehicle type-approved under Regulation 101 using the New European Driving Cycle (NEDC). The vehicle was equipped with a Li-ion battery with a capacity of 25 Ah and a nominal voltage of 345 V. PHEV had a 1.4 l.—110 kW gasoline engine. BEVx was a U.S. type-approved BEVx vehicle fitted with a Li-ion battery pack with a capacity of 60 Ah and a nominal voltage of 360 V. BEVx had a 0.65 l.—25 kW gasoline engine. Details of the tested vehicles are summarized in Table S1 (Supporting Information).

It should be noted that in the EU, a BEVx is considered, and tested, as any other OVC-HEV. However, according to the USA legislation, it falls under a different category than parallel PHEVs (i.e., OVC-HEV), namely, the BEVx category of the zero-emission vehicles.

2.2. Test Procedures. Emissions of regulated pollutants, ammonia (NH_3) and nitrous oxide (N_2O), as well as the vehicle electric range were studied at 23 °C and −7 °C. These temperatures were selected because 23 °C is the reference temperature used during the Type 1 test of the emission type-approval of LDVs in Europe, and −7 °C is the temperature used during the cold temperature procedure in different regions of the world (EU, USA, Korea, China).[21−24] Low temperature emission type-approval testing in EU (also known as Type 6 test) is performed without the use of the air heating system. However, to be more representative of the real use, the vehicles were also studied, during a second series of cold ambient temperature test (−7 °C), using the air conditioning system turned on and set at 21 °C (hereinafter −7 °C Aux-ON) as done by US EPA during the cold temperature vehicle testing.[24]

Figure S1 illustrates the experimental setup for the measurement of gaseous emissions and solid particle number (SPN). Regulated gaseous emissions were analyzed by sampling diluted exhaust from a set of Tedlar bags using an integrated system (MEXA-7400HTR-LE, HORIBA) equipped with a nondispersive infrared for CO and CO_2, a chemiluminescence for NOx, and a heated (191 °C) flame ionization detector for total hydrocarbons (THC). A solid particle number (SPN) measurement system (AVL APC 489), with particle diameter cut-off of 23 nm ($d_{50\%}$ = 23), compliant with the light-duty vehicles Regulation 83,[25] was used at the CVS to measure SPN (hereinafter PN). NH_3 and N_2O (unregulated pollutants) emissions were monitored from the raw exhaust using a high-resolution Fourier transform infrared spectrometer (FTIR; MKS 2030-HS). Further details on the

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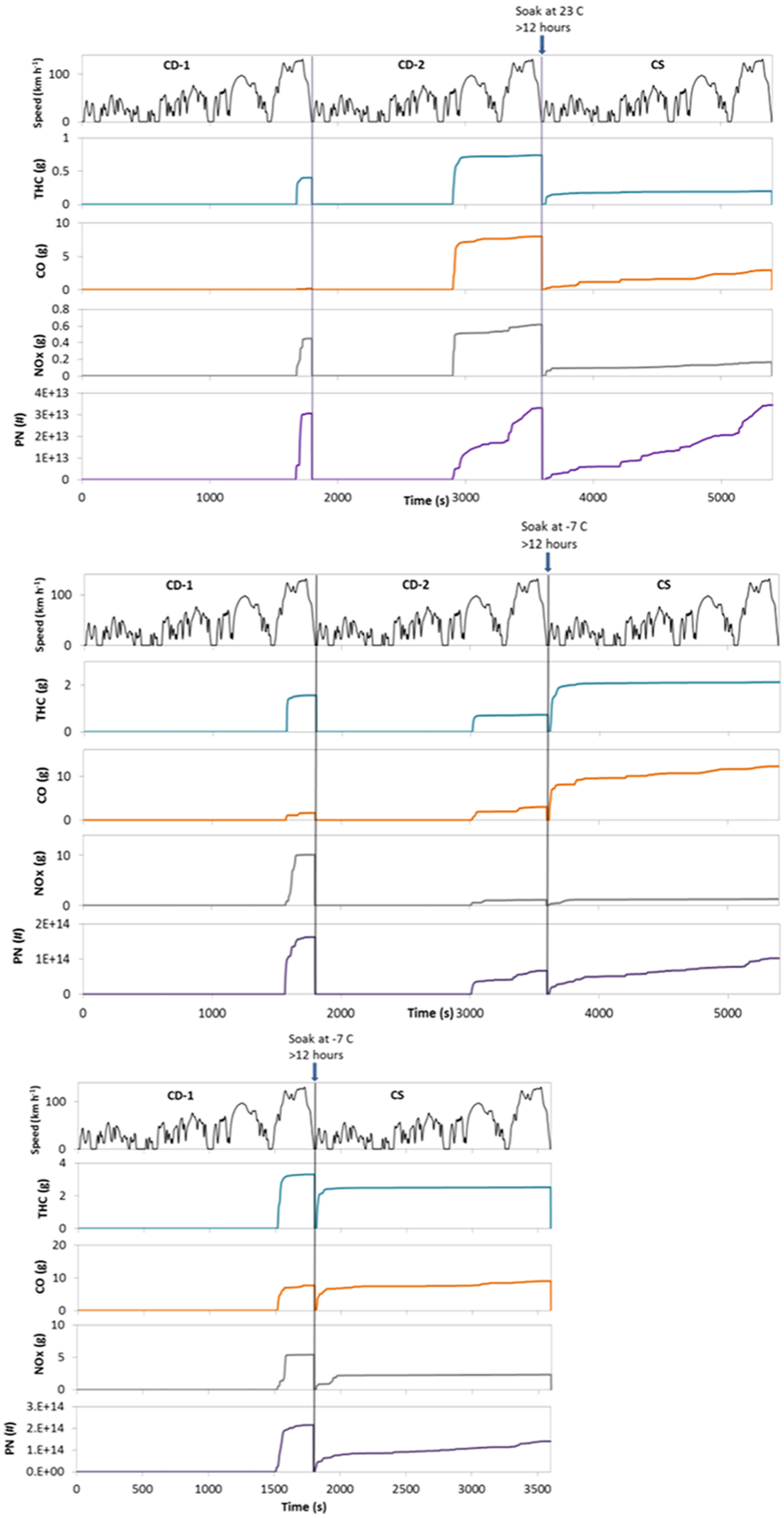

Figure 2. THC (blue line), CO (orange line), NOx (gray line), and PN (purple line) cumulative emissions from PHEV during charge-depleting (CD) and charge-sustaining (CS) sections of the WLTP type 1 test at 23 °C (top panels); −7 °C (central panels) and −7 °C ambient temperature using the vehicle's air conditioning system turned on and set at 21 °C (bottom panels).

performance during the real-time measurement on transient cycles using FTIR and emission factor calculations can be found in Suarez-Bertoa et al. (2014; 2015; 2017).[26−28] Calculations of the uncertainties of the measured pollutants were performed as described in Giechaskiel et al. (2012) and Giechaskiel et al. (2018).[29,30] Real-time battery voltage and

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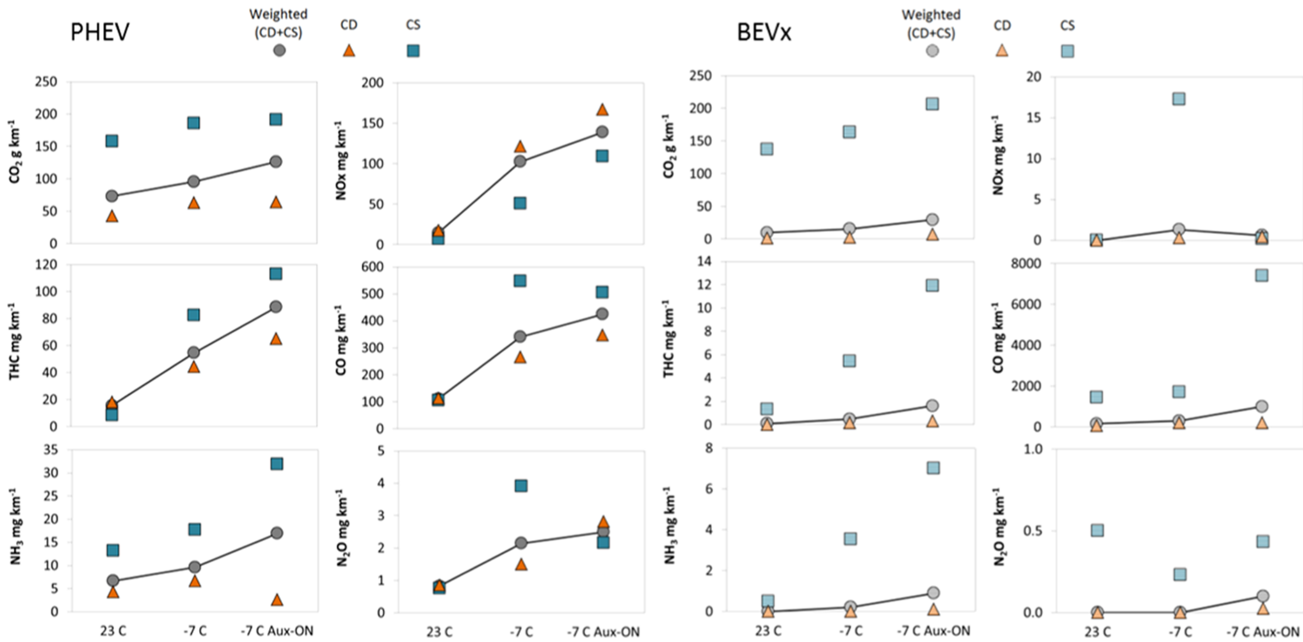

Figure 3. CO_2, NOx, THC, CO, N_2O, and NH_3 emission factors calculated during charge-depleting (CD) and charge-sustaining (CS) sections of the WLTP type 1 test as well as weighted emissions calculated weighting CD and CS using EU utility factors as described in GTR-15 at 23, −7, and −7 °C ambient temperature using the vehicle's air conditioning system turned on and set at 21 °C (−7 °C Aux-ON) for PHEV (left panel) and BEVx (right panel).

current were measured using a power analyzer (HIOKI 3390) and a data logger connected to the vehicle's engine control unit (ECU). Figures S2 and S3 illustrate the setup used to perform these measurements for PHEV and BEVx.

Vehicles were tested following option 3 of the test sequence for OVC-HEV described in Annex 8 of GTR-15.[31] A simplified scheme is shown in Figure 1. Hence, vehicles were tested in two different operation modes: (i) charge-depleting mode (CD), where the vehicle is tested in full electric operation, similar to a PEV; and (ii) charge-sustaining (CS) mode, where the ICE provides the energy that moves the vehicle and maintains a certain level of battery state of charge (SOC). CD testing comprises a number of WLTP tests carried out with a fully charged battery until break-off criterion (relative electric energy change (REEC) < 0.04) is reached (see GTR-15).[31] Once break-off criterion has been reached, the confirmation cycle starts and the vehicle is ready and preconditioned for the CS testing. CS test is a test performed following the procedures used for the conventional vehicles at cold-start conditions. During CD testing, the PHEV needed two worldwide harmonized light duty test cycle (WLTC) tests to reach the break-off criterion at 23 and −7 °C, and just one WLTC at −7 °C using the auxiliary systems (−7 °C Aux-ON). The BEVx needed six WLTC tests at 23 °C, five WLTC tests at −7 °C, and four WLTC at −7 °C using the auxiliary systems (−7 °C Aux-ON). Once the vehicles reached break-off criteria and the confirmation cycle was performed, they were soaked overnight at the required temperature. Then, they were tested in CS mode as prescribed in GTR-15.

To derive the CO_2 correction coefficients and correct CS CO_2 emissions to a neutral battery SOC level, five warm start CS tests were carried out with the same soaking time between them (30 min), as foreseen in the WLTP. Final vehicle emissions (also called weighted emissions) and electric range are calculated weighting the CD and CS operations (see eq 1 or refer to Annex 8 of GTR-15). To this aim, the so-called utility factors (UFs), which represent the ratio of the distance covered in the CD (electric) mode to the total distance covered between two subsequent charges, are used.

$$M_{i,\mathrm{weighted}}^{\mathrm{WLTP}} = \sum_{j=1}^{k} (\mathrm{UF}_j \times M_{i,\mathrm{CD},j}) + (1 - \sum_{j=1}^{k} \mathrm{UF}_j) \times M_{i,\mathrm{CS}} \quad (1)$$

where $M_{i,\mathrm{weighted}}$ is the utility factor-weighted mass emission of compound i (g/km), UF_j is the utility factor of WLTP's CD phase j, $M_{i,\mathrm{CD},j}$ is the pollutant mass emission of CD phase j (g/km), and $M_{i,\mathrm{CS}}$ is the charge-sustaining mass emission of gaseous emission compound i for the charge-sustaining Type 1 test (g/km). CD emissions of each phase j of the WLTP test (low, medium, high, and extra-high) have different weightings for the final CD emissions and will also weight differently if the WLTP test is the first or last in the CD sequence. Since phase-utility factors (UF_j) decrease with increase in the number of WLTP tests in the CD mode, the final CD emissions will decrease as the electric range increases. It should be noticed that before the introduction of the WLTP, NEDC CD emissions of CO_2 would equal 0 g/km if electric range of a vehicle was longer than 1 NEDC cycle (~11 km).

3. RESULTS AND DISCUSSION

A summary of the tests performed at the three studied conditions (i.e., 23, −7, and −7 °C Aux-ON) is illustrated in Figures 2 and 3 for PHEV and BEVx, respectively. Figure 3 shows that PHEV's ICE started working during the extra-high phase of the 1st cycle when the vehicle reached 130 km/h. However, PHEV was still capable of running in pure-electric mode for most part of the 2nd WLTC cycle. This particular behavior was discussed in a previous work.[32] There it was indicated that the energy management system of the vehicle may have been optimized for the NEDC, where the vehicle speed does not exceed 130 km/h. If the vehicle is prepared for

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Table 1. Electric Ranges and Energy Consumption of PHEV and BEVx Tested at Different Conditions[a]

	PHEV			BEVx		
	23 °C	−7 °C	−7 °C Aux-ON	23 °C	−7 °C	−7 °C Aux-ON
AER (km)	20.1	16.4	15.5	123.9	100.6	73.5
AER_city (km)	31.4	30.2	22.5	168.0	130.4	80.8
EEC weighed (Wh/km)	112	127	113	130	157	155
EEC CD (Wh/km)	152	173	221	138	171	174

[a]"−7 °C Aux-ON" refers to tests performed at −7 °C ambient temperature using the vehicle's air conditioning system turned on and set at 21 °C.

Table 2. Weighted Emission Factors of PHEV and BEVx Tested at Different Conditions[a]

	PHEV			BEVx		
	23 °C	−7 °C	−7 °C Aux-ON	23 °C	−7 °C	−7 °C Aux-ON
CO_2 (g/km)	73(±2)	95(±3)	126(±4)	10(±0)	15(±0)	29(±1)
THC	16(±2)	55(±6)	89(±9)	<1	1(±0)	2(±0)
CO	111(±11)	341(±34)	424(±42)	147(±30)	295(±30)	994(±99)
NOx	15(±3)	103(±4)	139(±6)	0	1(±1)	1(±1)
PN × 10^{11} (#/km)	12(±4)	44(±11)	64(±19)			
NH_3	7(±1)	20(±2)	17(±2)	0	<1	<1
N_2O	1(±1)	2(±1)	3(±0)	0	0	<1

[a]−7 °C Aux-ON refers to tests performed at −7 °C ambient temperature using the vehicle's air conditioning system turned on and set at 21 °C.

the testing under the WLTP, the combustion engine will not turn on during the first WLTC cycle and the electric ranges of this vehicle would be higher than the ranges shown in the next section (Table 1). A similar situation was seen when the vehicle was tested at −7 °C. During the test performed at −7 °C using the heating system, the battery was depleted much faster. In fact, ICE started before reaching the high-speed section of the extra-high phase of the 1st cycle. Hence, break-off criterion was met after just one WLTC.

3.1. Electric Range and Electric Energy Consumption.

Table 1 summarizes the electric ranges and electric energy consumption of PHEV and BEVx at three studied conditions (i.e., 23, −7, and −7 °C Aux-ON). All electric range (AER) is the distance traveled in pure-electric mode until the point when the ICE is turning on for the first time. AER_{city} is the electric range expected during the city driving only (low and medium speed phase of the WLTC). Weighted electric energy consumption (EEC weighed in Wh/km) is the total UF-weighted consumption where both driving modes (pure electric and hybrid) are considered. All these results, from tests done at 23 °C, must be communicated to the user through the vehicle's Certificate of Conformity (CoC). Table 1 also shows the values for UF-weighted electric energy consumption of the CD mode only (EEC CD) that is calculated and recorded during the vehicle's type-approval process, but not reported to the user in CoC.

BEVx has a higher C/M ratio (REESS capacity to vehicle Kerb mass ratio) compared to the PHEV, and hence, BEVx electric ranges were significantly higher than that of PHEV at all studied conditions. In good agreement with what was reported in previous studies,[11,15] electric ranges (AERs) dropped at low ambient temperatures and even more when the heating was used during testing. The BEVx's AER dropped from ∼124 km at 23 °C to ∼74 km at −7 °C (∼40%), and PHEV's AER dropped from ∼20 km at 23 °C to ∼15 km at −7 °C (∼25%) when the air heating system was used. Higher autonomy can be achieved when driving in city conditions (AER_{city}) compared to driving at higher speeds (high and extra-high speed phases) of the WLTC that represent the typical extraurban and highway conditions.

As a result of testing these vehicles at low ambient temperatures (−7 °C) and the use of heating, the electric energy consumption increased compared to the standard test conditions at 23 °C. In addition, the BEVx has a higher overall EEC weighed per km compared to the PHEV. Different results can be explained by the facts that EEC weighed is calculated by the formula where only 52% for PHEV and even 94% for BEVx of the CD EEC is considered for the overall contribution in the EEC (the remaining time the vehicles spend in CS modes). In addition, and since for the calculation of EEC weighed the number of CD tests is important, some numbers in Table 1 can be misleading to consumers, for example, lower values for −7 °C tests when the heating system was on compared to the values at −7 °C when the heating system was off. The EEC CD is a better parameter to compare the vehicle's electric energy consumption at different temperatures and with different electric loads. In addition, the EEC CD was higher for PHEV compared to the BEVx, and it closer depicts the conditions that a user can expect in terms of energy consumption when driving in pure-electric mode.

3.2. CO_2 Emissions.

Final emissions of CO_2 electrified vehicles are related not only to ICE size and performance, but also to the vehicle's electric range: high electric range is associated with lower CO_2 emissions (Tables 2, S2, and S3). Hence, PHEV presenting a lower electric range resulted in overall CO_2 emissions 5−7 times higher than BEVx.

As a consequence of UF-weighted calculation specified in the WLTP, the CD CO_2 emissions of PHEV were 8−27 times higher than BEVx CD CO_2 emissions at three different conditions (Table S2). CS CO_2 emissions were, as expected, higher for PHEV due to higher engine displacement and kerb mass. The only exception was found for CS tests at −7 °C (Table S3) using the heating system, where the CS CO_2 emissions of the BEVx were higher than the PHEV CS CO_2 emissions, suggesting a higher energy demand from the heating system in the case of the BEVx than for the PHEV. Moreover, under the WLTP procedure, the vehicle manufacturer has the

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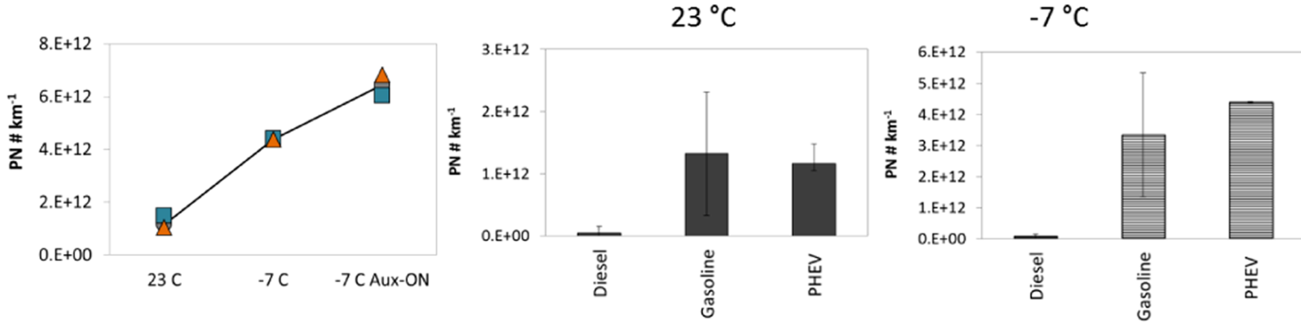

Figure 4. (Left panel) PN emission factors from PHEV calculated during charge-depleting (CD; orange triangle) and charge-sustaining (CS; blue square) sections of the WLTP Type 1 test as well as weighted emissions (gray circle) calculated weighing CD and CS using EU utility factors as described in GTR-15 at 23, −7, and −7 °C ambient temperature using the vehicle's air conditioning system turned on and set at 21 °C (−7 °C Aux-ON). (Right panel) Average PN emissions from conventional Euro 6 diesel and gasoline vehicles extracted from Suarez-Bertoa et al. (2017) compared to PHEV-weighted emissions at 23 °C (bottom-left panel) and −7 °C (bottom-right panel). In the case of diesel and gasoline vehicles, error bars represent the standard deviation of the values reported for the studied vehicles. PHEV's error bars represent the emission factor calculated from either the CD or CS tests.

possibility to correct the CS CO_2 emissions for the difference of the SOC of the battery between the start and end of the CS test. The PHEV CS CO_2 test with heating at −7 °C, which allowed this correction, resulted in a reduction of 31 gCO_2/km on the CS emissions initially measured (222 g/km). In the case of the BEVx CS CO_2 test with heating at −7 °C, the same correction resulted in reduction of 11 gCO_2/km on the CS emissions initially measured (217 g/km).

CO_2 emissions were strongly affected by the ambient temperature, which is in line with what was reported by Yuksel et al. (2016) and references therein.[11] In fact, the overall CO_2 emissions were, respectively, 30 and 61% higher for the PHEV and the BEVx at −7 °C than at 23 °C. This is partially due to the higher CO_2 emissions produced during the CS test at cold temperature but also because the pure-electric operation is shorter at −7 °C and therefore CD emissions were higher and CS emissions weigh more in the overall CO_2 emissions.

CO_2 emissions from the PHEV and the BEVx were, respectively, 72 and 209% higher when vehicles were tested at −7 °C and keeping the heating system on. Hence, the extra request of energy from the heating system led to a faster power consumption and consequently to the shorter pure-electric time operation.

CO_2 emissions from the PHEV were 3 times higher than those reported for a power split plug-in hybrid (AER ~83 km)[34] and 2 times higher than those for a series plug-in hybrid, both tested under similar conditions (23 °C using WLTP). The BEVx's CO_2 emissions were half of those reported by Badin et al.[34] These results indicate that a wide variety of OVC-HEVs is available and their electric range will be associated with the CO_2 that they will emit. The CO_2 final emissions from the PHEV and the BEVx were, respectively, 40% and 90% lower than the best performing Euro 6 conventional vehicle reported in a previous work.[33] The difference is even larger if only CD operation is taken into consideration. On the other hand, if only CS emissions were to be compared, CO_2 emissions from both vehicles were similar to what is reported in the literature for Euro 6 conventional vehicles.[33]

In this work, EU UFs have been used as prescribed by GTR-15. Plötz et al.[6] have shown that the real-world PHEV's CO_2 emissions and electric ranges can differ widely among users. They suggested that the main factors explaining this variation are the annual mileage, the regularity of daily driving, and the likelihood of long-distance trips. As Badin et al. recently reported,[34] higher emission factor would be expected if U.S. UFs are used in our study. As reported in WLTP regulation, once a significant number of PHEVs will be on the market, UF factors will have to be modified to be able to better capture their real usage and coherently assign the correct weighting to the values of CO_2 production.

The reported CO_2 emission factors refer to tailpipe emissions. These emissions are those declared by the manufacturers for the CO_2 fleet calculation and also used as information to the costumer. Hence, this approach does not reflect the emissions associated with the life cycle of the vehicle. A comprehensive comparison of emissions across vehicle types would be needed to account for the full life cycle, including emissions from power plants.

3.3. Regulated and Unregulated Emissions. Tables 2, S2, and S3 summarize the regulated and unregulated emission factors from the PHEV and the BEVx at all studied conditions (i.e., 23, −7, and −7 °C Aux-ON). Regulated emissions (with the exception of CO) and unregulated emissions were substantially higher for the PHEV vehicle with the lower C/M ratio than for the BEVx. CO emissions were higher for the BEVx in most cases (see Table 2).

Pollutant emissions disproportionally increased for the two studied vehicles as the ambient temperature decreased from 23 to −7 °C (see Figures 3 and 4). This behavior is similar to that previously reported for conventional vehicles.[33,34] To give an example, NOx-weighted combined emissions (i.e., UF-weighted CD + CS emissions) from PHEV were 7 times higher at −7 °C than at 23 °C. These differences were even larger when the vehicle's air heating system was used during the cold temperature tests (see Figures 3 and 4). Therefore, high pollutant emissions are expected from this type of vehicles under real driving conditions at cold ambient temperatures.

Plötz et al. recently reported[6] that a series of popular PHEVs are driven using exclusively the ICE for 22−61% of their operation time, which indicates that pure CS operation is not uncommon among PHEV drivers. The PHEV presented slightly higher pollutant emissions during CS (i.e., the vehicle uses the ICE, burning fuel, during most of the test) than during CD operation (i.e., the vehicle uses ICE during a fraction of the test sequence). The BEVx, on the other hand, presented substantially higher emissions during CS operation at all studied conditions (see Figure 3). During CS operation, the

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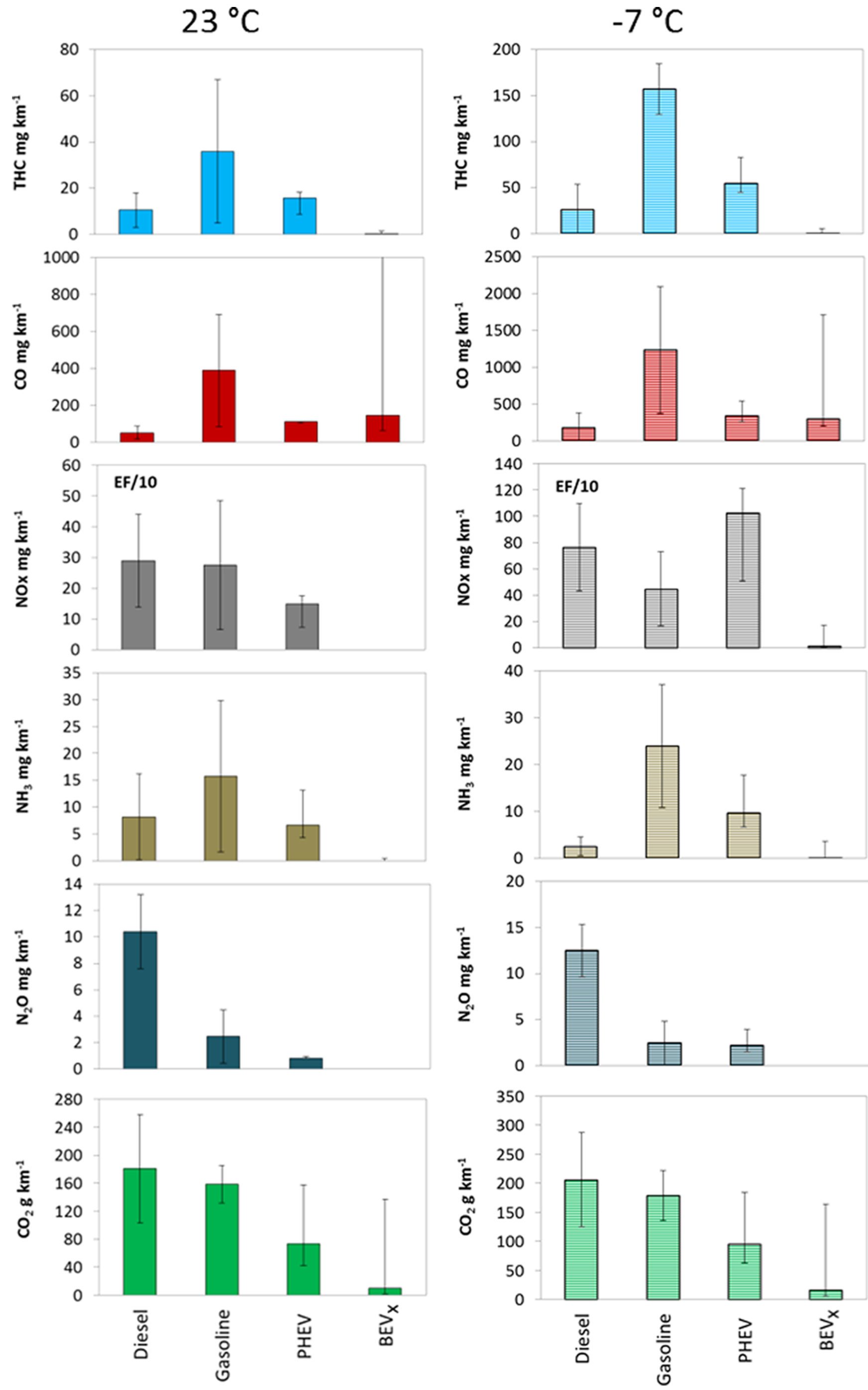

Figure 5. Average emissions from conventional Euro 6 diesel and gasoline vehicles extracted from Suarez-Bertoa et al. (2018) compared to PHEV- and BEVx-weighted emissions at 23 °C (left panel) and −7 °C (right panel). In the case of diesel and gasoline vehicles, error bars represent the standard deviation of the values reported for the studied vehicles. PHEV's and BEVx's error bars represent the emission factor calculated from either CD or CS tests. Diesel NOx emission factors are divided by 10 at the two temperatures.

PHEV emissions were, under all studied conditions, higher than those measured from the BEVx, with CO emissions being an exception.

The different emissions between the PHEV and the BEVx were, besides CO, mainly related to their corresponding electric ranges. The PHEV's electric range was relatively short (20 km); therefore, the weighting factor (UF) has less influence on the calculated CD emissions than in the case of long electric ranges. As a consequence, the PHEV's CD emissions were similar to those measured during the CS operation. The BEVx's electric range was considerably long, and for that reason, CD emissions were low. CS emissions are those obtained from a standard WLTP test with cold start, with the exception of CO_2 emissions, which can be corrected applying the procedure mentioned in the Experimental Section.

Since the PHEV's ICE started before the battery charge was depleted, the CO_2 emissions and range from the PHEV could be calculated considering a scenario where the ICE starts after ~40 km as shown by Pavlovic et al.[32] However, this approach

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cannot be applied to the other pollutants because the catalytic converter (and the engine) heats up when the ICE first started and the time elapsed until the second ICE ignition (that took place when the battery is actually depleted) was not enough to cool down the systems and again reach cold-start conditions. This scenario was more pronounced at cold temperature where the vehicle's engine temperature was 21 °C (instead of −7 °C cold start) when the ICE ignited during the second WLTC of the CD procedure.

BEVx presented very low emissions for most pollutants at all studied conditions (i.e., 23, −7, and −7 °C Aux-ON). Only BEVx's CO emissions were similar to those measured from PHEV and other conventional gasoline vehicles[33] (see Figure 5). The high energy demand during the highest speeds of the extra-high phase to the underdimensioned APU (650 cm^3 and 25 kW) could have led to the high CO emissions measured. At cold ambient temperature, the emissions during CS operation (vehicle uses the APU to charge a battery that is running low) were non-negligible. In fact, NOx emissions from BEVx during CS were comparable to those reported for conventional gasoline vehicles tested under similar conditions.[33] As previously indicated, it has been estimated that plug-in hybrids use up to 61% of their operation time in CS mode.[6] Hence, emissions from vehicles presenting a similar architecture should not be neglected in inventories and models.

PN emissions were measured for the PHEV (see the Experimental Section). Overall, the PHEV's PN emissions at 23 °C (1.2×10^{12} #/km) were above the values set for type-approval of Euro 6 vehicles (6×10^{11} #/km) using the NEDC (Table S1). PN emissions were 4 times higher (4.4×10^{12} #/km) during the tests at cold ambient temperature and 6 times higher (6.4×10^{12} #/km) when using the air heating system at −7 °C than those measured at 23 °C (1.2×10^{12} #/km) (Figure 2). Negative effects of cold ambient temperature on PN emissions from conventional gasoline vehicles have recently been reported.[33,36] Overall, PN emissions from the PHEV were similar to those reported from conventional gasoline vehicles and several orders of magnitude higher than those reported for modern (diesel particulate filter (DPF)-equipped) diesel vehicles (Figure 2).

The gaseous weighted emission factors from the PHEV were comparable to those previously reported[33] for a series of Euro 6 conventional passenger cars tested following the WLTP at 23 and −7 °C (see Figure 5). Nonetheless, emissions of NOx and N_2O from conventional diesel vehicles and THCs from conventional gasoline vehicles at −7 °C, reported in Suarez-Bertoa and Astorga (2018), were higher than those from PHEV. On the other hand, THC, CO, and NH_3 emissions from the PHEV at −7 °C were higher than those reported for diesel vehicles studied under similar conditions by Suarez-Bertoa and Astorga (2018).[33] Moreover, NOx emissions from the PHEV at −7 °C were 25% higher than the worse gasoline vehicle reported in that study.[33] Emission factors on the CS mode were, in most cases, higher than the obtained weighted emission factors (which also account for mileage in pure-electric driving). It is then evident that if driven with a depleted battery, the PHEV would result in even higher emissions compared to conventional vehicles. As illustrated in Figures 3 and 4, cold ambient temperatures would further exacerbate this difference.

Holland et al.[15] have reported that whereas gasoline-vehicle-related damages are large in Los Angeles due to the large population and properties of the airshed, PEVs' damages are small due to the clean Western US power grid, resulting in substantial environmental benefits for the PEVs. However, due to the prevalence of coal-fired generation and cold ambient temperatures, PEVs' damages are large in the upper Midwest. In their study, Holland et al. do not integrate the effect of temperature on conventional gasoline vehicles as they consider it to have a small effect. However, it has been shown that emissions of CO_2 and other pollutants considered in Holland's analysis dramatically increase at cold ambient temperatures. We found that pollutant emissions of the studied vehicles can be as high as those measured from gasoline vehicles at 23 and −7 °C and that at cold ambient temperature the electric range reduction is in line with what Holland et al. considered in their analysis for PEVs. Hence, it appears important to include plug-in hybrid vehicles in this kind of holistic analysis as they could play a negative role in the two areas described by Holland et al., that is, a similar role to that of gasoline vehicles in Los Angeles, and the role played by a combination of conventional gasoline and PEV in the upper Midwest, having even higher negative impact in these cold regions.

Recent studies reported that cold-start emissions of VOCs, CO, and NOx, often taking place during the first 30−60 s for modern petrol vehicles,[37] account for most of the total emissions of conventional spark ignition vehicles.[35] These emissions have also been reported to largely increase at cold ambient temperatures.[35,38,39] On the CS mode, the THC, CO, and NOx cold-start emissions from PHEV were the main fraction of the total emissions at all the studied conditions (see Figure 2). Furthermore, the cold-start emissions during the CD mode were even higher than those registered during the CS mode. In fact, total THC, NOx, and PN emissions reached approximately the same levels during the few seconds that the ICE was used in the 1st CD cycle as those produced during the entire CS cycle (see Figure S2).

Such high pollutant emissions may result from the catalyst conditioning strategy of the manufacturer (also known as "cat-heating" event),[40] which for this vehicle took place during the extra-high speed phase of the 1st CD test. In the worst case scenario, one could expect emissions to be similar to those observed during a CS test as in both cases the ICE ignites from a cold-start condition and with a cold catalytic converter. However, as illustrated in Figure 2, ignition of the ICE during high engine loads during the CD mode resulted in disproportionally high emissions each time the ICE suddenly ignited during transient operation. This effect was more accentuated at cold ambient temperatures, where the pollutants that were emitted during the few seconds of ICE operation of the 1st CD cycle were several orders of magnitude higher than during the whole cold-start CS test.

These results put together highlight that in the few seconds of ICE use, current plug-in hybrid vehicles could emit as many particles, THC, and NOx as those emitted by Euro 6 conventional gasoline vehicles during the cold-start or even during an entire WLTP test (23 km and 30 min long) and several times more if running in cold ambient temperatures. Moreover, the PN emissions would be much larger than those typically measured from modern (DPF-equipped) diesel vehicles.

The levels of THC and NH_3 emissions measured suggest that vehicles like the PHEV will contribute as much as an average conventional vehicle to the formation of atmospheric secondary organic and inorganic aerosol.[38,41] These secondary aerosols are a major air-quality concern as they account for

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most of the PM found in urban environments.[42,43] Moreover, with the NOx emissions measured, these vehicles do not appear to be a valid solution to reduce urban NOx levels.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsomega.8b02459.

Vehicle specifications; experimental setup for the measurement of the pollutant emissions and for the measurement of current and voltage from the tested vehicles; charge-depleting and charge-sustaining emission factors; THC, CO, and NOx cumulative emissions from BEV (PDF)

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Notes

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ABBREVIATIONS

AER, all electric range
APU, auxiliary power unit
BEVx, range-extended battery electric vehicle
EC-JRC, European Commission Joint Research Centre
ECU, engine control unit
CD, charge-depleting mode
EEA, European Environmental Agency
CS, charge-sustaining mode
EU, European Union
GHG, greenhouse gases
GTR, global technical regulations
ICE, internal combustion engine
LDVs, light-duty vehicles
OVC-HEV, off-vehicle charge hybrid electric vehicles
PEV, pure-electric vehicles
PHEV, plug-in hybrid electric vehicle
REEC, relative electric energy change
REESS, rechargeable electric energy storage system
SOC, state of charge
SPN, solid particle number
SULEV, super ultralow emission vehicle
TZEV, transitional zero-emission vehicles
UF, utility factor
UNECE, United Nations Economic Commission for Europe
VELA, Vehicle Emission Laboratories
WLTP, world harmonized light-duty test procedure

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